



Simultaneous nitrification–denitrification for the treatment of high-strength nitrogen in hypersaline wastewater by aerobic granular sludge



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ABSTRACT

Fish processing industries produce wastewater containing high amounts of salt, organic matter and nitrogen. Biological treatment of such wastewaters could be problematic due to inhibitory effects exerted by high salinity levels. In detail, high salt concentrations lead to the accumulation of nitrite due to the inhibition of nitrite-oxidizing bacteria. The feasibility of performing simultaneous nitrification and denitrification in the treatment of fish canning wastewater by aerobic granular sludge was evaluated, and simultaneous nitrification–denitrification was successfully sustained at salinities up to 50 gNaCl L⁻¹, with a yield of over 90%. The total nitrogen concentration in the effluent was less than 10 mg L⁻¹ at salinities up to 50 gNaCl L⁻¹. Nitrification collapsed above 50 gNaCl L⁻¹, and then, the only nitrogen removal mechanism was represented by heterotrophic synthesis. In contrast, organic matter removal was not affected by salinity but was instead affected by the organic loading rate (OLR). Both COD and BOD removal efficiencies were over 90%. The COD fractionation analysis indicated that aerobic granules were able to remove more than 95% of the particulate organic matter. Finally, results obtained in this work noted that aerobic granular sludge had an excellent ability to adapt under adverse environmental conditions.

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1. Introduction

Fish processing industries require a huge amount of water throughout all production steps, including fish cleaning, cooling, sanitization and floor washing (Cristovao et al., 2015). Depending on the fish to be processed, large amounts of salt in the form of brine are used for obtaining the finished products. Therefore, wastewaters generated by fish factories are characterized by high salt (mainly sodium chloride) concentrations and extreme variability, depending on the process carried out. Nevertheless, fish canning wastewaters are characterized by high suspended solids (up to 5000 mg L⁻¹), organic matter (COD up to 90,000 mg L⁻¹) and nitrogen concentrations (TN up to 3000 mg L⁻¹), especially during periods of fresh fish processing, due to the presence of blood, entrails, oil, flakes and fish tissues (Chowdhury et al., 2010). Due to the high salinity level, their biological treatment would require the application of halophilic microorganisms, which are able to tolerate

high salt concentrations. In contrast, non-halophilic bacteria are not suitable for this purpose because of cell disintegration due to the osmotic pressure difference across the cellular membrane that causes the loss of cellular water (plasmolysis) (Dincer and Kargi, 2001). However, the application of non-halophilic microorganisms is possible because a moderate acclimation of activated sludge to high salinity has been tested (Lefebvre and Moletta, 2006; Di Bella et al., 2015).

Currently, aerobic granular sludge is considered to be one of the most promising biological wastewater treatment technologies, thanks to high settling velocity, high biomass retention, simultaneous removal of carbon and nitrogen and their ability to withstand high organic load (Long et al., 2015). For these unique properties, aerobic granular sludge has attracted increasing interest for industrial wastewater treatment (Zhang et al., 2011; Abdullah et al., 2013). Among these, aerobic granular sludge reactors have been successfully used for treating synthetic saline wastewater. Taheri et al. (2012) obtained stable aerobic granules at 10 gNaCl L⁻¹, whereas Wang et al. (2015) successfully adapted aerobic granular sludge at higher salinity levels of up to 80 gNaCl L⁻¹. The authors

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found that some microorganisms adapting to high salinity gradually became the predominant bacteria. However, all of these studies noted remarkable issues regarding nitrogen removal. Bassin et al. (2011) stated that ammonia-oxidizing bacteria (AOB) could tolerate salt concentrations of up to 33 gNaCl L⁻¹, and Pronk et al. (2014) observed that nitrite-oxidizing bacteria (NOB) showed complete inhibition at 20 gNaCl L⁻¹. Wan et al. (2014) found that under NaCl concentrations of up to 50 gNaCl L⁻¹, full partial nitrification can be achieved, and the nitrite accumulation rate reached nearly 100%. Consequently, at high salt concentrations, the accumulation of either ammonia or nitrite usually occurs when treating saline wastewater. In any case, these studies reported that when treating synthetic saline wastewater, the total nitrogen removal efficiency was moderate and was rarely higher than 70%. Apart from these studies, few experiences with real saline wastewater have been carried out. Figueroa et al. (2008) treated fish canning effluent characterized by salt contents of up to 30 gNaCl L⁻¹. Although organic carbon was completely depleted, nitrogen removal efficiencies were lower than 40%. As in other cases, the main issue was the insufficient nitrogen removal efficiency. Similar results were obtained by Val del Río et al. (2013), who also observed significant disintegration of granules for organic load rates (OLR) higher than 4.4 kgCOD m⁻³d⁻¹. Because high amounts of nitrite accumulation commonly occur in the treatment of saline wastewater, simultaneous nitrification–denitrification processes could be effectively coupled with granular sludge technology to improve nitrogen removal. The simultaneous nitrification–denitrification process has been successfully applied for the treatment of synthetic wastewater by means of aerobic granular sludge (Lochmatter et al., 2014; Li-long et al., 2014). However, to date, this solution has not been applied for treating real wastewater, so it needs to be investigated. Moreover, as reported by De Kreuk et al. (2010), aerobic granular sludge is able to remove particulate substrates. Because the particulate substrate is an important fraction of the organic material in fish canning wastewater (Jemli et al., 2015), aerobic granular sludge reactors could be a good solution for their biological treatment.

The main goal of this study was to analyse the feasibility of treating fish canning wastewater by using aerobic granular sludge in a sequencing batch airlift reactor (SBAR) (Beun et al., 2002). In detail, the study of nitrogen removal by means of nitrification–denitrification processes at high salinity was tested at various salt concentrations. For this purpose, a 78-day experiment was carried out with the goal of finding the best operating conditions for maximizing reactor performance, evaluated in terms of nitrogen and organic carbon removal, at increasing salt concentrations (from 30 gNaCl L⁻¹ up to 75 gNaCl L⁻¹). Special attention has been paid to the maximum salt concentration that allows satisfactory nitrogen removal efficiency to be attained.

2. Materials and methods

2.1. Reactor and experimental set-up

The reactor was operating for 78 days divided into 5 periods characterized by different salt concentrations and organic load rates. The reactor was a column-type (100 cm height) with a working volume of 3.5 L (internal diameter of 8.6 cm) and was characterized by an internal riser 50 cm high with an internal diameter of 5.4 cm. Air was introduced via a fine bubble aerator at the base of the reactor at a flow rate of 3 L min⁻¹ so that the hydraulic shear forces were approximately 2.4 cm s⁻¹. The filling height was 70 cm, so the height/diameter ratio was equal to 7. The effluent was discharged by a solenoid valve placed at 35 cm from the base of the reactor. Thus, the volumetric exchange ratio (VER)

was 50% for each cycle. The experimentation was divided into five periods, each of which was characterized by a different salt concentration. The raw wastewater (salinity equal to 150 gNaCl L⁻¹) was the same for the whole experiment, and it was diluted with tap water to obtain different salt concentrations in each sub-period. From periods I to IVa, the salt concentrations increased from 30 gNaCl L⁻¹ (I) to 38 gNaCl L⁻¹ (II), 50 gNaCl L⁻¹ (III) and 75 gNaCl L⁻¹ (IVa). From periods I to IVa, the reactor was operating on a 12 h per cycle, included 45 min of influent feeding, 665 min of aeration, 5 min of settling and 5 min of effluent withdrawal. To reduce the organic load rate in period IVb without changing the influent salinity, the cycle length was extended to 24 h, keeping each phase duration constant excepted for the aeration phase. A Programmable Logic Controller (PLC) automatically handled the SBR cycling operations. Due to the long reaction cycle, organic carbon was almost completely degraded before the end of the cycle. Certainly, this limited the availability of the organic substrate for nitrite reduction. Thus, to supply an electron donor for nitrite reduction, a known amount of water spiked sodium acetate (200 mg L⁻¹) was added as an external carbon source from the second part of period II (from the 10th hour of the cycle) to enhance denitrification without interrupting aeration. Due to the simultaneous presence of dissolved oxygen that was preferentially used by heterotrophic bacteria as an electron acceptor for acetate oxidation instead of nitrite, the acetate dose was increased to 400 mgL⁻¹ in period III. The sludge retention time was maintained at 45–50 days by daily purging a known amount of mixed liquor volume.

2.2. Wastewater characterization

The fish canning wastewater was collected from a local industry that produces canned anchovies. The fish canning production process begins with the arrival of fresh fish (fish processing phase). Thus, heads and viscera are removed and the fishes are stored in brine. During this process, water is enriched in blood, oil, flakes and salt. Consequently, wastewaters are characterized by very high organic matter content (COD 80000 mg L⁻¹), high total suspended solids concentrations (4621 mg L⁻¹) and high salinity levels (up to 300 gNaCl L⁻¹). After a period of storage (approximately 1–2 months), fish pass to the canning section in which they are washed, preserved in tins and packaged for distribution. The salt and organic matter contents of the process wastewater remain high but are significantly lower than those of the previous phase (COD = 16,000 mg L⁻¹ and NaCl = 25 mg L⁻¹). Due to the extreme variability of wastewater composition, in order to analyse the biological performance of wastewaters having nearly the same characteristics, a large volume of raw sewage was collected at one time during the fish-processing phase and stored at 4 °C for the whole experimental period. The main characteristics of the raw wastewater are reported in Table 1.

Hereafter, to obtain the desired salinity, wastewater was diluted with tap water in accordance with a dilution factor ranging from 2 to 5 (v/v). The duration of each phase was not fixed a priori. The phases were changed when steady state conditions, in terms of nutrient removal efficiency, were reached. Each phase lasted at least 15 days. The organic load rate was not controlled, but it was proportional to the dilution factor applied. The main operating conditions are reported in Table 2.

2.3. Analytical methods

All of the chemical–physical analyses (COD, BOD, NH₄–N, NO₃–N, NO₂–N, TSS, and VSS) were performed according to standard methods (APHA, 2005). Soluble COD was determined after filtration through a 0.45 µm membrane; hence, particulate COD

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